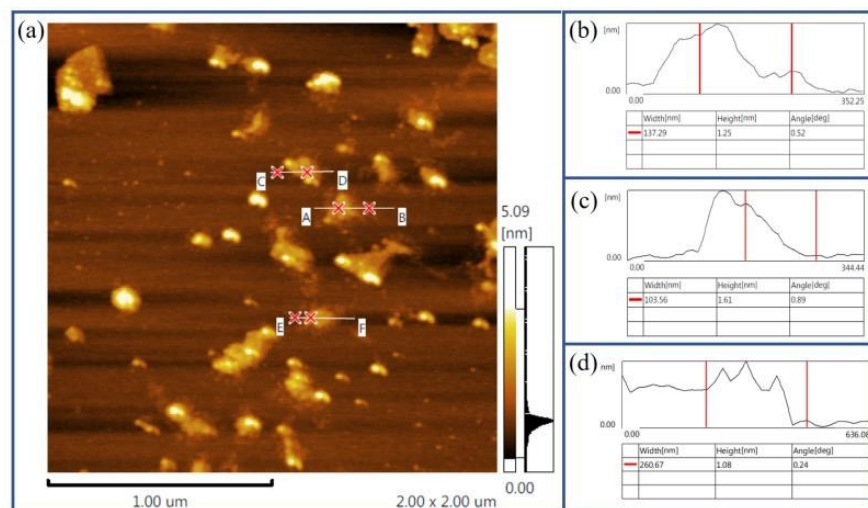


## Electrochemical Investigation of Adsorption of Graphene Oxide at an Interface between Two Immiscible Electrolyte Solutions

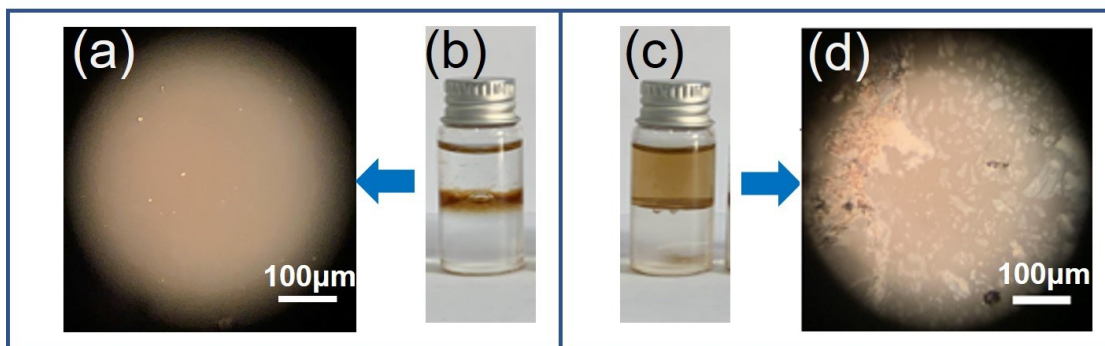
Haiyan Qiu,<sup>a</sup> Tao Jiang,<sup>a</sup> Xiaoyuan Wang,<sup>a</sup> Lin Zhu,<sup>a</sup> Qingwei Wang,<sup>a</sup> Yun Zhao,<sup>a</sup> Jianjian Ge<sup>b</sup> and Yong Chen<sup>\*a</sup>

<sup>a</sup> School of Chemical and Environmental Engineering, Shanghai Institute of Technology, Shanghai 201418, China

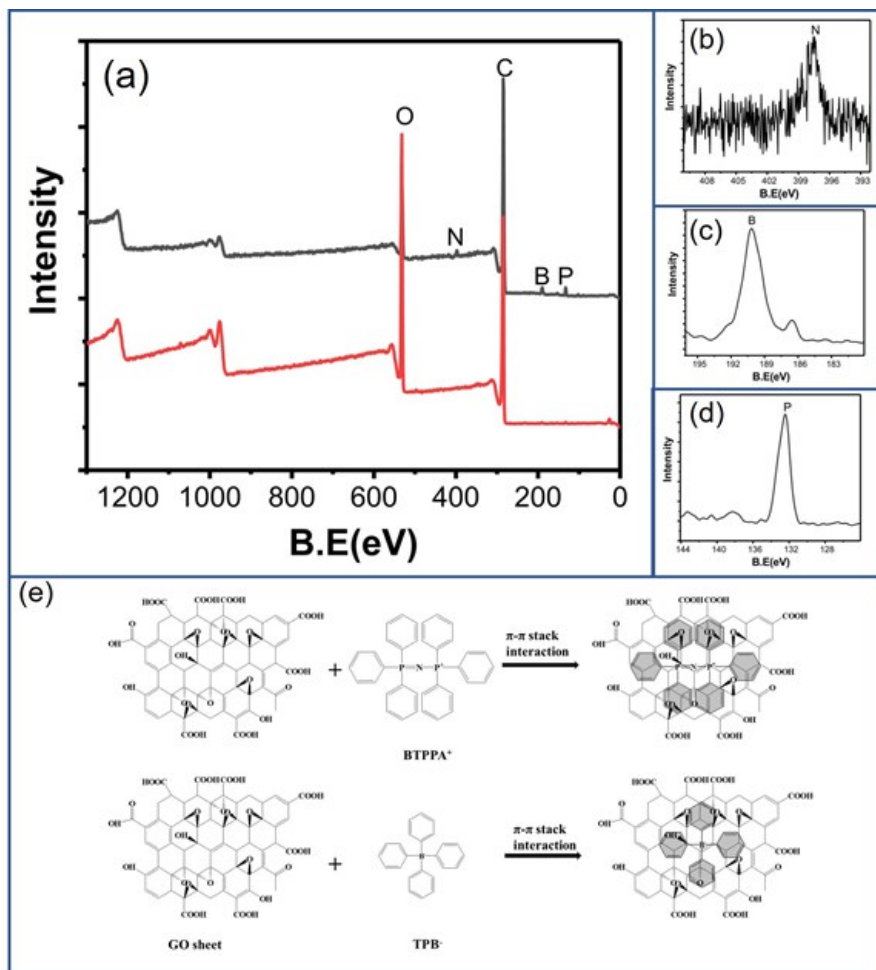
<sup>b</sup> School of Science, Shanghai Institute of Technology, Shanghai 201418, China.



**Fig. S1** AFM image (a) and the height analyses (b-d) of graphene oxide nanosheets. The GO nanosheets were characterized as an average size of 50 to 200 nm and a height of  $1.31 \pm 0.02$  nm.



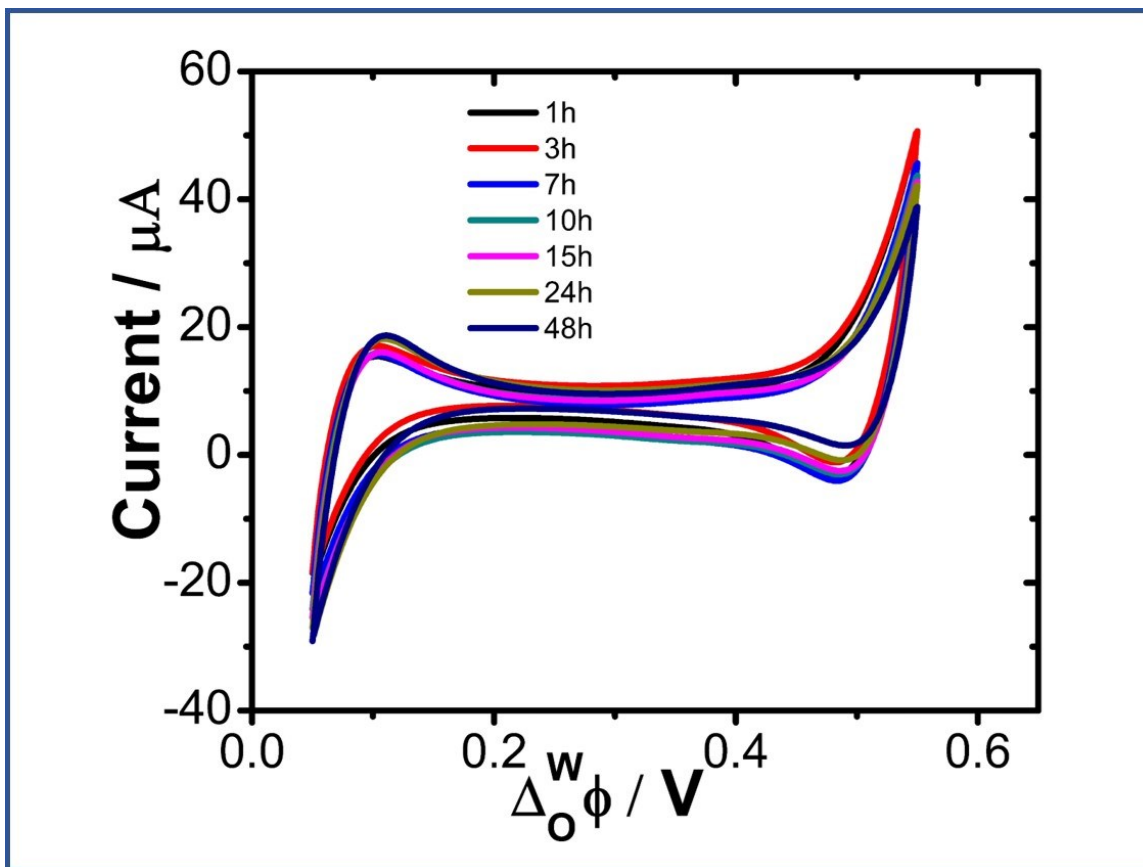
**Figure. S2** The top-viewed optical microscopy (a) and the side-viewed photograph (b) for the adsorption of GO at the W/DCE interface if aqueous phase contains the supporting electrolyte LiCl and organic phase contains no electrolyte BTPPATPB; The side-viewed photograph (c) and the top-viewed optical microscopy (d) for the adsorption of GO at the W/DCE interface if organic phase contains the supporting electrolyte BTPPATPB and aqueous phase contains no electrolyte LiCl.



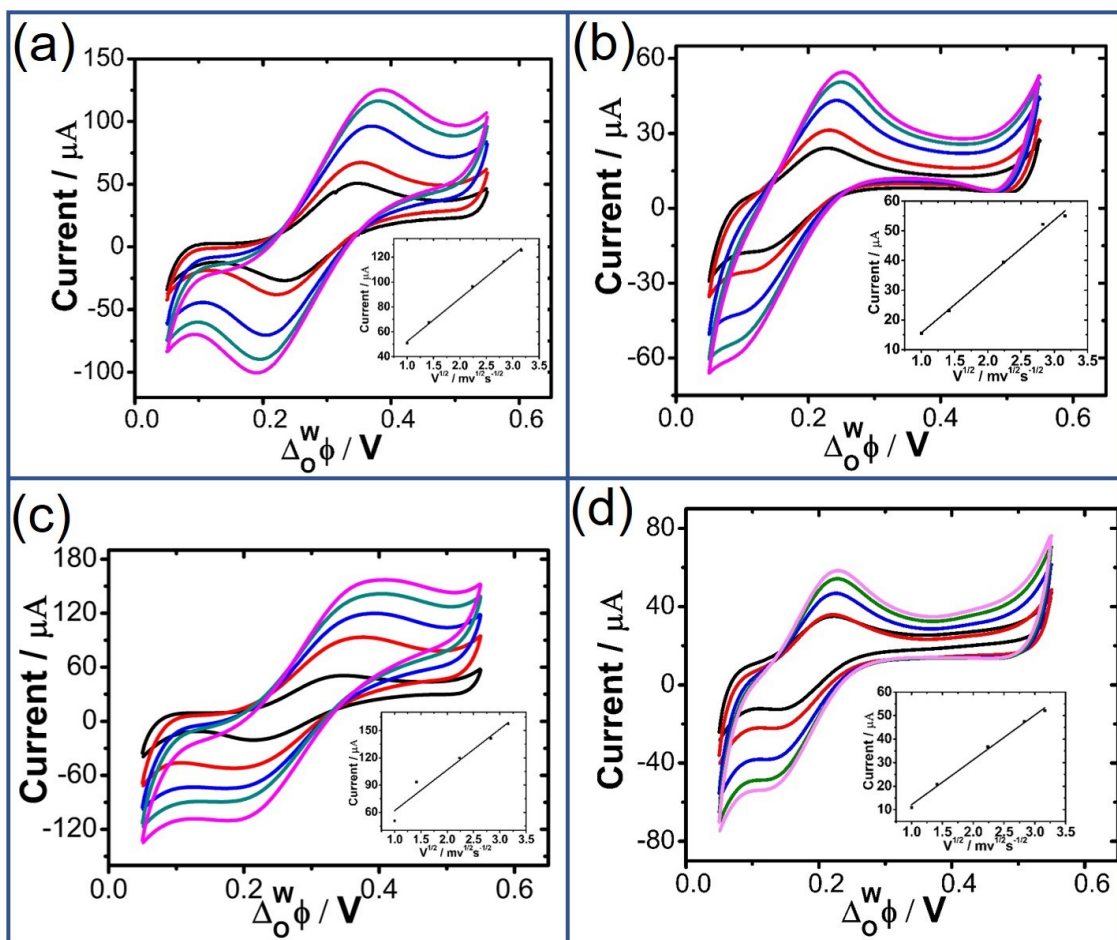
**Figure. S3** XPS survey spectra (a-d) obtained for the GO layers extracted from the water/DCE interface and (e) the scheme for the  $\pi$ - $\pi$  interaction between the residual conjugate domains in GO and the aromatic ions of organic electrolyte, BTPPA<sup>+</sup> and TPB<sup>+</sup>. The red line in (a) is the XPS survey spectrum of pure GO. The sample preparation method for the XPS measurement of GO layers is described as follows: after the experiment of GO adsorption at the W/DCE interface, the upper aqueous phase containing the interfacial GO layers and the possible DCE solution adsorbed on the GO layers was sucked out using a straw and putted into a separatory funnel. After that, 2 ml pure DCE was added into the separatory funnel, which was shaken slowly 1 min. After left it standing still about 10 mins for the complete separation between aqueous phase and DCE phase, the lower DEC phase was released from the lower part of the separatory funnel. Repeated such an operation three times in order to overcome the effect of DCE solution on the measurement of XPS. Subsequently, the aqueous solution containing GO layers was filtrated to obtain GO layers, which was washed by deionized water. Such a process was repeated for three times to remove the aqueous electrolyte LiCl. Finally, as-obtained GO layers were dried in oven under 60 °C for 12 hours. Then, a GO sample was prepared for the XPS measurement.

Element Sample	C1s /%	O1s /%	B1s /%	N1s /%	P2p /%
GO <sub>W/DCE</sub>	69.42	26.08	1.74	1.76	1.00
GO <sub>Pure</sub>	70.30	29.70	-	-	-

**Table. S1** The element contents of pure GO and the GO layers extracted from the W/DCE interface calculated from their XPS spectra.



**Figure. S4** The changes of CVs with time in the presence of GO (0.1 g L<sup>-1</sup>) in aqueous phase (cell 1) at a scan rate of 5 mV/s.



**Figure. S5** CVs for the ion transfer of both  $\text{TEA}^+$  (a,c) and  $\text{ClO}_4^-$  (b,d) across the GO-modified W/DCE interface under  $\text{pH} \sim 7.4$  (a, b) and  $\sim 5.4$  (c, d). The insets in each figure are the corresponding linear relationships between the peak current for the ion transfer of each ion from W to DCE and the square root of the different scan rates (1, 2, 5, 8, 10 mV/s).

Parameters	$\text{pH} \sim 7.4$		$\text{pH} \sim 5.4$	
	$I_p / \mu\text{A}$	$D_w / \text{cm}^2 \text{s}^{-1}$	$I_p / \mu\text{A}$	$D_w / \text{cm}^2 \text{s}^{-1}$
$\text{TEA}^+$	$58.78 \pm 0.04$	$(1.18 \pm 0.02) \times 10^{-5}$	$55.85 \pm 0.05$	$(1.15 \pm 0.02) \times 10^{-5}$
$\text{ClO}_4^-$	$48.89 \pm 0.03$	$(1.09 \pm 0.01) \times 10^{-5}$	$43.27 \pm 0.04$	$(1.03 \pm 0.02) \times 10^{-5}$

**Table. S2** The electrochemical parameters obtained for the ion transfer of  $\text{TEA}^+$  and  $\text{ClO}_4^-$  across the GO-layers-modified W/DCE interface under  $\text{pH} \sim 7.4$  and  $\sim 5.4$ , including the peak currents of the ion transfer of each ion from W to DCE ( $I_p$ ) at a scan rate of 5 mV/s and the diffusion coefficients ( $D_w$ ) of each ion in the aqueous phase evaluated from Fig. S5.